Lecture 4. Optical properties cont'd

1. Extinction by an ensemble of particles

$$\frac{I}{I_0} = \exp(-\varepsilon_{ext} z) = \exp(-\tau)$$

where $\tau = \varepsilon_{ext} z$ is the **aerosol optical depth**, and I_0 is the incident intensity.

The **extinction coefficient** of N particles of diameter D (monodisperse distribution), refractive index m, and at wavelength λ is

$$\varepsilon_{ext}(\lambda) = \frac{\pi D^2}{4} N Q_{ext}(m, x)$$

with population of different sized particles, ε_{ext} is given by

$$\varepsilon_{ext}(\lambda) = \int_0^{D_p^{max}} \frac{\pi D_p^2}{4} Q_{ext}(m, x) n(D_p) dD_p \ ,$$

where D_p^{max} is the upper limit of particle diameter.

The extinction can be expressed as the sum of the scattering and absorption coefficients: $\varepsilon_{ext} = \varepsilon_{scat} + \varepsilon_{abs}$

Furthermore, we can decompose ε_{scat} and ε_{abs} into contributions from the gas and particle components of the atmosphere:

$\varepsilon_{scat} = \varepsilon_{sg} + \varepsilon_{sp}$ where ε_{sg} is the scattering coefficient due to gases (Rayleigh scattering) and ε_{sp} is the scattering coefficient due to particles.

$\varepsilon_{abs} = \varepsilon_{ag} + \varepsilon_{ap}$ where ε_{ag} is the absorption coefficient due to gases

and ε_{ap} is the absorption coefficient due to particles.

By using the mass distribution function, $n_M(D_p) = \rho_p \dfrac{\pi D_p^3}{6} n(D_p)$, and substituting $n(D_p)$ with it in the expression of the extinction coefficient ε_{ext}, we obtain

$$\varepsilon_{ext}(\lambda) = \int_0^{D_p^{max}} \frac{3}{2\rho_p D_p} Q_{ext}(m,x) n(D_p) dD_p$$

which can be expressed as $\varepsilon_{ext}(\lambda) = \displaystyle\int_0^{D_p^{max}} E_{ext}(m,\lambda,D_p) n(D_p) dD_p$ where
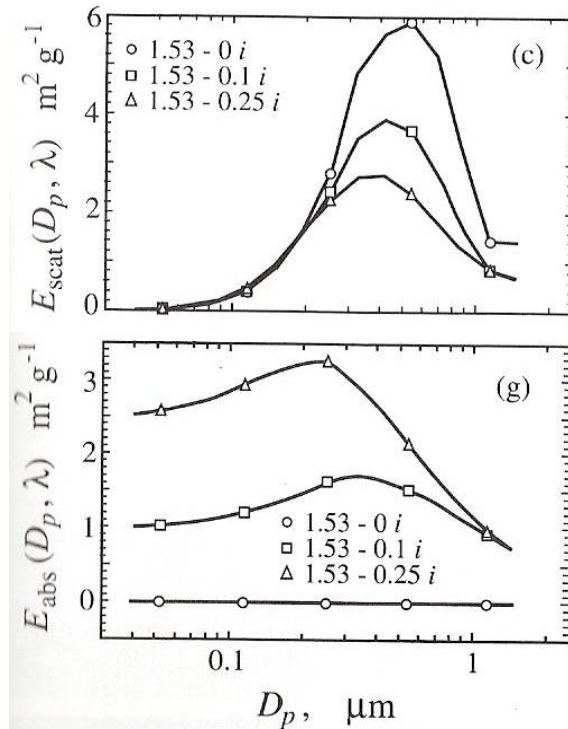
$$E_{ext}(m,\lambda,D_p) = \frac{3}{2\rho_p D_p} Q_{ext}(m,x) \text{ is the } \textbf{mass extinction efficiency}.$$

Similarly, the **mass scattering** and **mass absorption efficiencies** are

$$E_{scat}(m,\lambda,D_p) = \frac{3}{2\rho_p D_p} Q_{scat}(m,x)$$

$$E_{abs}(m,\lambda,D_p) = \frac{3}{2\rho_p D_p} Q_{abs}(m,x)$$

Figure 4.1. Mass scattering and absorption efficiencies for materials having refractive indices $m=n+ik$, with $n=1.53$ and $k=-0, -0.1, -0.25$ (Seinfeld and Pandis, 1998).

Assuming a spatially uniform aerosol layer of depth h with a number size distribution $n(D)$. The aerosol optical depth is given by $\tau = \varepsilon_{ext} h$ and ε_{ext} is calculated by integration the mass extinction over all radii. As a first approximation, the aerosol size distribution can be approximated as monodisperse with a number concentration N and an effective radius r_e, and $\tau = hN\pi r_e^2 Q_{ext}$. Knowing the aerosol concentration $m = \frac{4}{3}\pi r_e^3 \rho_p N$ of density ρ_p, the aerosol optical depth can be easily calculated from the simple formula:

$$\tau = \frac{3Q_{ext} m h}{4r_e \rho_p} = \frac{3Q_{ext} M}{4r_e \rho_p}$$

where M is the vertically integrated mass column [kg.m^{-2}].

Table 4.1. Particle density, effective radius, extinction efficiency for dry aerosols.

Aerosol type	Density [g.cm^{-3}]	R$_e$ [μm]	Q$_{ext}$ at 550nm
Sulfate	1.7	0.156	1.34
OC	1.8	0.087	0.68
BC	1	0.039	0.56
Dust	2.6	0.14	1.3
	2.6	0.24	2.2
	2.6	0.45	2.67
	2.6	0.8	2.68
	2.6	1.4	2.42
	2.6	2.4	2.28
	2.6	4.5	2.18
Sea salt	2.2	0.8	2.7
	2.2	5.7	2.1

Hygroscopic aerosols:

Certain aerosols are able to take up water vapor from the atmosphere snd grow in volume, process known as hygroscopicity. If the amount of water taken up is sufficient to form spontaneously a solution, the aerosol (salt) is said to deliquesce. In general the crystallization of the salt is taking place a lower RH than the deliquescence.

Figure 4.2. Particle size change as a function of relative humidity for sulfate aerosols at 25°C (Tang et al., 1978).
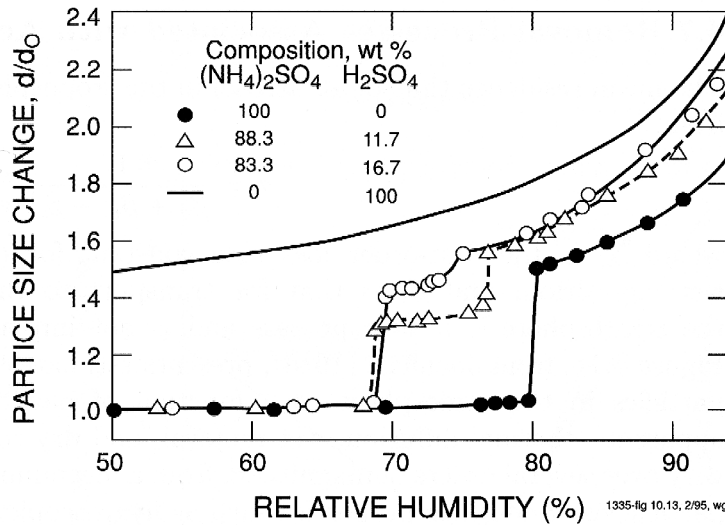
In case of wetted aerosols, the most convenient way of linking the aerosol optical depth and the aerosol dry mass M_d is to express τ as $\tau = \beta M_d$ where β is given by:

$$\beta = \frac{3 Q_{ext} M}{4 \rho r_e M_d}$$, so that all the humidification effects are embodied in β.

The density of the hygroscopic particle is evaluated as $\rho = f_d \rho_d + (1 - f_d) \rho_w$ with f_d being the volume fraction of dry aerosol within an aerosol particle, and ρ_w the density of water.

Table 4.2. Hygroscopic growth factors of $r_e/r_{e,dry}$ at different RH (from *Chin et al., 2001*)

RH (%)	0	50	70	80	90	95	99
Sulfate	1	1.4	1.5	1.6	1.8	1.9	3.5
OC	1	1.2	1.4	1.5	1.6	1.8	2.2
Sea salt	1	1.6	1.8	2	2.4	2.9	4.9

Mixture of aerosols:

When the atmosphere is composed of aerosol types of different refractive indices and size distribution, the mass extinction efficiency, single scattering albedo and asymmetry parameter are calculated for each aerosol types. Assuming a mixture of n aerosol types with all particles externally mixed, the optical depth is given by:

$$\tau = \sum_{i=1}^{n} \tau^i = \sum_{i=1}^{n} E_{ext}^i M_{tot}^i$$

Where E_{ext}^i is the mass extinction efficiency of component i [m^{2}g^{-1}], and M_{tot}^i is the mass column of component i [gm^{-2}]

The single scattering albedo of the mixture is calculated by weigthing the single scattering albedo of each component by its corresponding optical depth:

$$\omega = \sum_{i=1}^{n} \frac{\omega^i \tau^i}{\tau}$$

And the asymmetry parameter is calculated similarly by weighting each components value by the optical depth

$$g = \sum_{i=1}^{n} \frac{g^i \tau^i}{\tau}$$

For internally mixed aerosols, there are several approaches (called mixing rules) to calculate an effective refractive index m_e of the internally mixed aerosols:

1. Volume (or mass) weighted mixing:

    $m_e = \sum_i m_i f_i$, where m_i is the refractive index of component i and f_i is its volume fraction.

2. Bruggeman approximation for two randomly mixed species, provides the effective refractive index from the following relation

    $$f_1 \frac{\varepsilon_1 - \varepsilon_e}{\varepsilon_1 + 2\varepsilon_e} + f_2 \frac{\varepsilon_2 - \varepsilon_e}{\varepsilon_2 + 2\varepsilon_e} = 0$$

    Where ε_i is the dielectric constant (recall $m = \sqrt{\varepsilon}$) of component i and f_i is the volume fraction

3. Maxwell-Garnett approximation for two species when one is a matrix (host material) with a dielectric constant ε_2 and the other is an inclusion of dielectric ε_1:

$$f_1 \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2} = \frac{\varepsilon_e - \varepsilon_2}{\varepsilon_e + 2\varepsilon_2}$$

$$f_2 \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + 2\varepsilon_1} = \frac{\varepsilon_e - \varepsilon_1}{\varepsilon_e + 2\varepsilon_1}$$

The Mie theory is for homogeneous spheres, but in most cases particles are of different shapes and configurations. The exact calculations of Mie theory can also be applied to:

1. Layered spheres
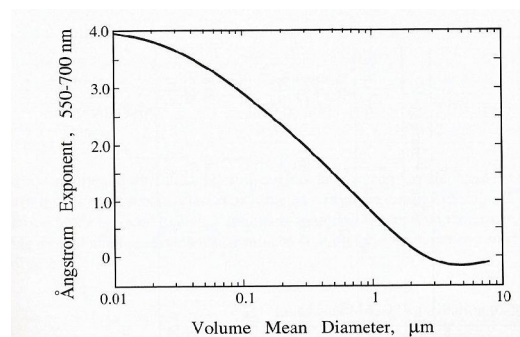2. Infinite cylinders
3. Layers of infinite cylinders

Useful properties

Co-albedo: $1 - \omega$

Absorption optical depth: $\tau_a = (1 - \omega)\tau$

Angstrom exponent: $\alpha = -\dfrac{d \log \varepsilon_{ext}}{d \log \lambda} \cong -\dfrac{\log(\frac{\varepsilon_{ext1}}{\varepsilon_{ext2}})}{\log(\frac{\lambda_1}{\lambda_2})} = -\dfrac{\log(\frac{\tau_1}{\tau_2})}{\log(\frac{\lambda_1}{\lambda_2})}$

Figure 4.3 Angstrom exponent for a log-normal distribution of water aerosol (σ_g =2) with refractive index m=1.33-0i in the wavelength range 550 to 700 nm (Seinfeld and Pandis, 1998).


Angstrom exponent (computed from t measurements on two different wavelengths) can be used to find t on another wavelength using the relation

$$\tau_2 = \tau_1(\lambda_1 / \lambda_2)^\alpha$$

Upscatter fraction: The fraction of light that is scattered by a particle into the upward hemisphere relative to the local horizon, and can be calculated by integrating the angular distribution of light intensity scattered by the particle.

Upscatter fraction

$$\beta = \frac{\int_{\pi}^{2\pi} P(\Theta)\sin\Theta\, d\Theta}{2\int_0^{\pi} P(\theta)\sin\theta\, d\theta}$$
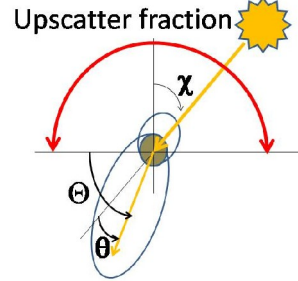
where $\Theta = \dfrac{\pi}{2} + \theta - \chi$, χ is the solar zenith angle, and $P(\theta)$ is the scattering phase function. When the sun is at nadir, $\chi = 0$, $\Theta = \dfrac{\pi}{2} + \theta$ and the upscatter fraction β corresponds to the hemispheric backscatter ratio b (also called simply backscatter ratio).

$$b = \frac{\int_{\pi/2}^{\pi} P(\theta)\sin\theta\, d\theta}{\int_0^{\pi} P(\theta)\sin\theta\, d\theta}$$

Scattering effect of an aerosol Layer

The fraction of a solar beam transmitted through an aerosol layer of optical depth τ is $e^{-\tau}$ and the fraction reflected back in the direction of the beam is $r = (1 - e^{-\tau})\omega\beta$. The fraction of light absorbed within the layer is $(1 - \omega)(1 - e^{-\tau})$. The fraction scattered downward is $\omega(1 - \beta)(1 - e^{-\tau})$. The total fraction of radiation incident on the layer that is transmitted downward is $t = e^{-\tau} + \omega(1 - \beta)(1 - e^{-\tau})$. For a surface albedo R_s, the incident radiation that is reflected is $R_s t$. The reflected radiation will be partly transmitted (fraction $t^2 R_s$) and partly scattered back to the Earth's surface (fraction trR_s). The beam reflected downward is itself reflected off the surface, and the process is repeated. The fraction of incident light which is ultimately reflected back to the top of the atmosphere (TOA) is $r + t^2 R_s (1 + R_s r + (R_s r)^2 + (R_s r)^3 + ...)$.
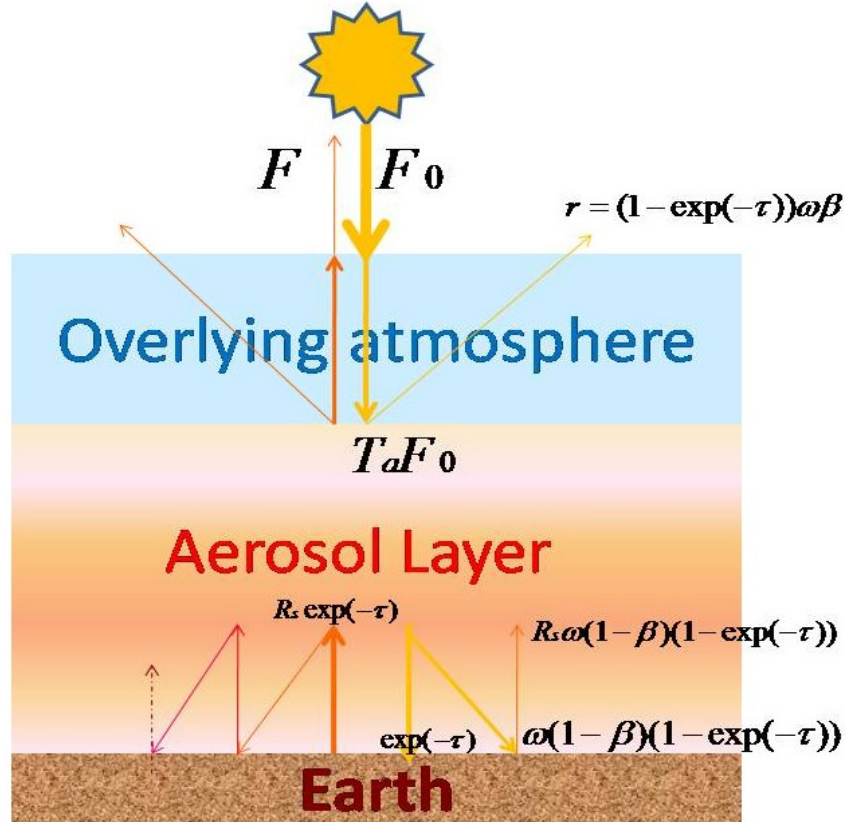
By summing the series of $\dfrac{1}{1 - R_s r}$, the total upward flux is

$$F_r = (r + \frac{t^2 R_s}{1 - R_s r})F_0$$

The term in parenthesis is the total reflectance R_{as} of the aerosol-surface system, and the change of reflectance due to the aerosol layer ΔR_p is given by the relation:

$$\Delta R_p = (r + \frac{t^2 R_s}{1 - R_s r}) - R_s$$

Which depends on :

1. Single scattering albedo ω
2. Aerosol optical depth τ
3. Upscatter fraction of the aerosol β
4. Albedo of the Earth's surface (also called planetary albedo) R_s

The total reflectance depends also on clouds cover (A_c) and molecular scattering. Let T_a denote the fractional transmittance of the atmosphere. The change in global average planetary albedo associated with aerosol layer is described as

$$\Delta R_p = T_a^2 (1 - A_c) \left[(r + \frac{t^2 R_s}{1 - R_s r}) - R_s \right]$$

Table 4.3. *Global mean values and their uncertainty range (from IPCC Third Assessment Report, 2001, accessible at http://www.grida.no/climate/ipcc_tar/wg1/197.htm).*

The optical parameters are for wavelength of 550nm and are for dry aerosol.

Quantity	Central value	Range
Aerosol mass scattering efficiency E_{sca} [$m^2.g^{-1}$]	3.5	2.3 to 4.7
Aerosol single scattering albedo ω	0.92	0.85 to 0.97
Atmospheric transmittance above aerosol layer T_a	0.87	0.72 to 1
Fractional increase in aerosol scattering efficiency due to hygroscopic growth at RH=80%	2	1.7 to 2.3
Fraction of Earth not covered by cloud	0.39	0.35 to 0.43
Mean surface albedo	0.15	0.08 to 0.22

2. Haze and Visibility.

Clean (background) atmospheric conditions: light is scattered and absorbed by natural gases and particulates (background aerosol).

Polluted atmospheric conditions: air pollutants (gases and particles) cause additional attenuation of light.

Haze is a form of air pollution consisting of small particles of dust, soot, sulfates, and other material.

- Haze has natural and anthropogenic sources.

Total suspended particulate (TSP) refers to the total mass concentration of aerosol particles present in the air.

- In heavily polluted cities, average TSP abundance is about 50-100 $\mu m/m^3$, with upper limits of about 1000 $\mu m/m^3$.

Two major problems caused by haze:

1. visibility reduction;
2. health effects

Visibility is generally used synonymously with "visual range", meaning the farthest distance at which one can see a large, black object against the sky at the horizon.

Some factors determining how far one can see through the atmosphere:

i.   optical properties of the atmosphere;
ii. amount and distribution of light;
iii. characteristics of the objects observed;
iv. properties of the human eye.

- Visibility is reduced by the absorption and scattering of light by both gases and particles. However, light scattering by particles is the most important phenomenon responsible for visibility degradation.

Clean (background) atmospheric conditions: one can see over distances up to several hundred kilometers.

Polluted atmospheric conditions: visibility is up to 10 km.

Koschmieder equation:

relates visual range (visibility), x_v, and extinction coefficient ,ε_{ext}, as

$$x_v = 3.912/\varepsilon_{ext}$$

NOTE: in Koschmieder equation the extinction coefficient is sum of extinction coefficients of all gases and particles, which attenuate light.

NOTE: in Koschmieder equation the extinction coefficient is averaged over visible wavelengths, however it is often taken at about 550-nm wavelength